# $E$-band excitations in the magnetic Keplerate molecule $Fe_{30}$

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The low-temperature excitations in the magnetic Keplerate molecule $Fe_{30}$ as calculated by linear spin-wave theory (SWT), modified linear SWT, and spin-level mean-field theory (SLMFT), are compared to the recent inelastic neutron scattering results by Garlea et al. [Phys. Rev. B **73**, 024414 (2006)]. SLMFT reproduces a part of the experimental spectrum rather well, but not all of it. SWTs yield a small fraction of the $E$-band excitations and hence are not capable of a complete description of the excitation spectrum.

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The subject of the elementary spin excitations in finite antiferromagnetic (AFM) Heisenberg spin clusters has become of much interest recently, as for many experimentally available molecular nanomagnets, such as the AFM wheels or the Keplerate molecule $Fe_{30}$, the Heisenberg spin Hamiltonian

$$\hat{H} = -\sum_{i,j} J_{ij} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \tag{1}$$

is the appropriate starting point for a discussion.[1,2] $J_{ij}$ measures the exchange interaction between spins $i$ and $j$, and $S_i$ is the length of spin $i$. The number of spin centers in the cluster will be denoted as $N$.

For a number of AFM Heisenberg clusters, the low-lying energy spectrum exhibits a rather particular structure: As function of total spin $S$, the lowest-lying levels form a set of rotational bands (RBs) for which $E(S) \propto S(S+1)$.[3,4,5,6] The lowest set of bands is denoted as $L$ band; the higher-lying ones as $E$ band (and the remaining states as quasi continuum).[6] The $L$ band is related to a quantized rotation of the Néel vector. For bipartite systems, the $E$ band corresponds to (discrete) AFM spin-wave excitations. This picture of the excitations has been confirmed experimentally for a number of bipartite systems;[2] in particular the AFM wheels.[7]

Also for the molecule $Fe_{30}$ a RB structure was conjectured.[8] This system consists of an icosidodecahedral arrangement of 30 spin-5/2 centers and is characterized by three AFM sublattices.[9] For the existence of the $L$ band in this molecule solid evidence from both theory and experiment is available.[1,10] Concerning the $E$ band, however, the situation is less clear. This brief report is motivated by two recent studies, a study of the excitations using modified spin-wave theory,[11] and an inelastic neutron scattering (INS) experiment, where the observed scattering intensity was related to the $E$-band excitations.[12] In the following, the spin excitations in $Fe_{30}$ as calculated by linear spin-wave theory (LSWT), modified linear spin-wave theory (mLSWT), and spin level mean-field theory (SLMFT), as well as observed experimentally by INS are compared.

For the SWT calculations, Ref. [11] was closely followed, albeit with a different numerical implementation. The classical ground state of $\hat{H}$ (Ref. 13) is determined by the conditions $\mathbf{S}_i \times \sum_j J_{ij}\mathbf{S}_j = 0$ for each $i$,[14] which are solved numerically by iterating $\mathbf{S}_i = S_i \sum_j J_{ij}\mathbf{S}_j / |\sum_j J_{ij}\mathbf{S}_j|$ until convergence.[15] Local coordinate frames are introduced at each spin site such that the local $z$-axes coincide with the directions of the classical spins. Writing $\tilde{\mathbf{S}}_i = (\hat{S}_i^+, \hat{S}_i^-, \hat{S}_i^z)^T$ for the spherical spin operators in the local frames, the Hamiltonian becomes

$$\hat{H} = -\sum_{ij} J_{ij} \tilde{\mathbf{S}}_i \cdot \mathbf{U}_{ij} \cdot \tilde{\mathbf{S}}_j. \tag{2}$$

Expressions for the elements $U_{ij}^{\nu\mu}$ ($\nu, \mu = +, -, z$) are given in [16]. The spin operators (in the new frames) are expressed by Holstein-Primakoff bosons, $\hat{S}_i^+ \approx \sqrt{2S_i} a_i$, $\hat{S}_i^z = S_i - a_i^\dagger a_i$, yielding $\hat{H}_B = -\sum_{ij} J_{ij} \hat{C}_{ij}^{(2)}$ with

$$\begin{aligned}\hat{C}_{ij}^{(2)} &= (S_i S_j - S_j a_i^\dagger a_i - S_i a_j^\dagger a_j) U_{ij}^{zz} + 2\sqrt{S_i S_j} \\ &\quad \times (U_{ij}^{++} a_i a_j + U_{ij}^{--} a_i^\dagger a_j^\dagger + U_{ij}^{+-} a_i a_j^\dagger + U_{ij}^{-+} a_i^\dagger a_j).\end{aligned}$$

This real-space bosonic Hamiltonian is Bogliubov diagonalized numerically. We follow Ref. [17] and introduce $\mathbf{a}^\dagger = (a_1^\dagger, a_2^\dagger, \ldots, a_N^\dagger, a_1, a_2, \ldots, a_N)$, yielding

$$\hat{H}_B = E_0 + \mathbf{a}^\dagger \cdot \mathbf{D} \cdot \mathbf{a}, \tag{3}$$

with the $2N \times 2N$ matrix

$$\mathbf{D} = \begin{pmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{B}^* & \mathbf{A}^* \end{pmatrix} \tag{4}$$

and the $N \times N$ matrices $\mathbf{A}$ and $\mathbf{B}$, which depend on $S_i$, $J_{ij}$ and $U_{ij}^{\nu\mu}$. The properties $\mathbf{A}^\dagger = \mathbf{A}$ and $\mathbf{B}^T = \mathbf{B}$ ensure the hermiticy of $\mathbf{D}$, and $\hat{H}_B$. The Bogliubov diagonalization (or, in the language of [17], para-diagonalization) introduces $N$ new bosons $c_k$ via $\mathbf{c}^\dagger = \mathbf{a}^\dagger \mathbf{V}^\dagger$. The $2N \times 2N$ matrix $\mathbf{V}$ obeys $\mathbf{V}^\dagger \check{\mathbf{1}} \mathbf{V} = \check{\mathbf{1}}$ with $\check{\mathbf{1}} = \mathrm{diag}(\mathbf{1}, \mathbf{-1})$ ($\mathbf{1}$ is the $N \times N$ unit matrix). This ensures the boson character of the $c_k$. Para-diagonalization corresponds to finding a $\mathbf{V}$ with $(\mathbf{V}^\dagger)^{-1} \cdot \mathbf{D} \cdot \mathbf{V}^{-1} = \frac{1}{2}\mathbf{E}$, where $\mathbf{E} = \mathrm{diag}(E_1, E_2, \ldots, E_N, E_1, E_2, \ldots, E_N)$. The para-diagonalized Hamiltonian then reads

$$\hat{H}_B = E_0 + \sum_k E_k \left( c_k^\dagger c_k + \frac{1}{2} \right). \tag{5}$$

Numerical algorithms for determining $\mathbf{E}$ and $\mathbf{V}$ are given in [17]. The procedure so far corresponds to linear SWT (LSWT), i.e., $\hat{H}_B^{(LSWT)} \equiv \hat{H}_B$. The ground-state and excitation energies are given by $E_0^{(LSWT)} = E_0 + \frac{1}{2}\sum_k E_k$ and $E_k^{(LSWT)} = E_k$, respectively.

Standard SWTs, such as LSWT, start from the assumption of an ordered, symmetry-broken ground state, which is obviously incorrect for finite clusters. In the modified SWTs,[11,18,19] spin-rotational invariance is restored "by hand" by enforcing zero on-site magnetizations, $\langle \hat{S}_i^z \rangle = S_i - \langle a_i^\dagger a_i \rangle = 0$ ($\langle . \rangle$ denotes the quantum expectation value in the ground state). $N$ Lagrange multipliers $\mu_i$ are introduced, and the boson Hamiltonian of modified linear SWT (mLSWT) becomes

$$\hat{H}_B^{(mLSWT)} = \hat{H}_B + \sum_i \mu_i (a_i^\dagger a_i - S_i). \qquad (6)$$

With $A'_{ij} = A_{ij} + (\mu_i/2)\delta_{ij}$ and $E'_0 = E_0 - \sum_i \mu_i (S_i + \frac{1}{2})$, also $\hat{H}_B^{(mLSWT)}$ assumes the form of Eq. (3). Para-diagonalization yields $\hat{H}_B^{(mLSWT)} = E'_0 + \sum_k E'_k (c_k'^\dagger c'_k + \frac{1}{2})$. The ground-state and excitation energies, $E_0^{(mLSWT)} = E'_0 + \frac{1}{2}\sum_k E'_k$ and $E_k^{(mLSWT)} = E'_k$, implicitly depend on the Langrange multipliers, which are found numerically by solving $\hat{H}_B^{(mLSWT)}$ iteratively until $\langle a_i^\dagger a_i \rangle = S_i$ for each $i$.

A further approach, called spin-level mean-field theory (SLMFT), was explored. The RB structure assumed for $Fe_{30}$ implies approximating the wavefunctions of the $L$ band by the spin levels $|\alpha S_A S_B S_C S M\rangle$, where $S_A$, $S_B$, and $S_C$ are the total spins of the three AFM sublattices, and $S_A = S_B = S_C = S_i N/3$ ($\alpha$ denotes intermediate spin quantum numbers).[4,6,8,20] This is equivalent to describing the $L$ band by the three-sublattice Hamiltonian $\hat{H}_{ABC} = -6J/N(\hat{\mathbf{S}}_A \cdot \hat{\mathbf{S}}_B + \hat{\mathbf{S}}_B \cdot \hat{\mathbf{S}}_C + \hat{\mathbf{S}}_A \cdot \hat{\mathbf{S}}_C)$.

The $E$ band, which corresponds to excitations out of the $L$ band, is then related to the spin levels with $S_A = S_B = S_i N/3$, $S_C = S_i N/3 - 1$, and the permutations thereof.[6,8,20] These states are degenerate for $\hat{H}_{ABC}$, but are expected to be split by the perturbation $\hat{H} - \hat{H}_{ABC}$. This suggests, as a first approximation, to diagonalize $\hat{H}$ in the subspace of the spin levels $|\alpha S_A S_B S_C S M\rangle$, with $S_A$, $S_B$, and $S_C$ chosen as appropriate for the $L$ and $E$ bands. In this work, where only the zero-temperature excitation spectrum is considered, $\hat{H}$ was diagonalized in the subspace of the $S$=0 ground state and the $S$=1 states of the $L$ and $E$ bands.[21] According to the coupling rules of three spins, the $S$=0 ground state is non-degenerate, and the $S$=1 sectors of the $L$ and $E$ band consist of 3 and 81 spin levels, respectively. The numerical calculation of the matrix elements $\langle \alpha S_A S_B S_C S M | \hat{H} | \alpha S_A S_B S_C S M \rangle$ is simple thanks to the irreducible tensor operator techniques.[22]

Sublattice Hamiltonians, such as $\hat{H}_{ABC}$, may be "derived" by Fourier analysis of the full Hamiltonian.[3,4,20] This procedure works for spin clusters with large symmetry, such as rings, polyhedrons, or extended lattices. It may be constructed also by replacing each spin by the mean-field spin $\hat{\mathbf{S}}_\eta = 1/N_\eta \sum_{i\in\eta} \hat{\mathbf{S}}_i$ of its corresponding sublattice $\eta$, which provides a more general method.[23]

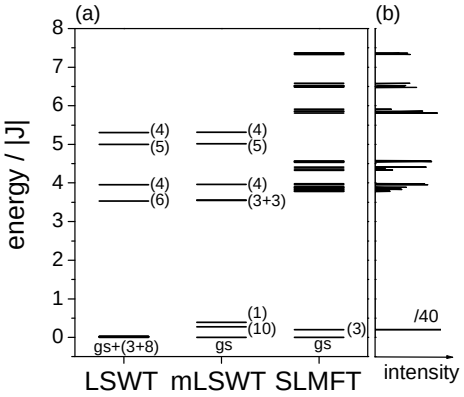


FIG. 1: (a) Energy levels of $Fe_{30}$ as calculated by LSWT, mLSWT, and SLMFT (gs = ground state; numbers in brackets give the degeneracy). (b) $T = 0$ neutron scattering intensity calculated by SLMFT. The intensity of the peak at $0.2|J|$ was divided by a factor of 40.

The results of the calculations for the excitations in $Fe_{30}$, as obtained by LSWT, mLSWT, and SLMFT, are displayed in Fig. 1(a) (the mLSWT results were obtained previously in [11]). In LSWT, the excitation spectrum consists of 3 levels at zero energy (with respect to the ground state), followed by 8 levels in the energy range 0.0078 to $0.0247|J|$, and four bands in the energy range $3.5-5.5|J|$. As expected, mLSWT strongly affects the 11 lowest excitations, the spectrum is in particular gapped. The first 10 excitations are at $0.2758|J|$, followed by one at $0.3900|J|$. The higher-lying excitations are affected by less than 0.3% (the levels at ca. $3.5|J|$ are weakly split into two subgroups). Clearly, the 11 low-lying excitations, and the ground-state, should be related to the $L$ band, while the remaining higher-lying excitations should be related to the $E$ band. SLMFT produces a more structured energy spectrum. It consists of the three triplets of the $L$ band at $0.2|J|$, and the 81 triplets of the $E$ band, which span the range 3.7811 to $7.3682|J|$. For comparison, $\hat{H}_{ABC}$ gives energies of 0.2 and $5.2|J|$ for the $L$ and $E$ band, respectively.

All three theories yield a significant splitting of the $E$ band, which is about $2|J|$ for the SWTs and $3.5|J|$ for the SLMFT. The experimental INS data at 65 mK, which is reproduced in Fig. 2, in fact shows a broad feature from 0.2 to 1.1 meV. Within the three-sublattice Hamiltonian approach it was modeled by a single Gaussian of width 0.66 meV.[12] Its position was determined to 0.56 meV, which corresponds to $J = -0.108$ meV, in rough agreement with the value $J = -0.134$ meV inferred from magnetic susceptibility.[1,12] The above theories partially account for the observed broadening (as noted before for the SWTs in [11,12]). Since SLMFT produces the largest splitting, it is compared to experiment

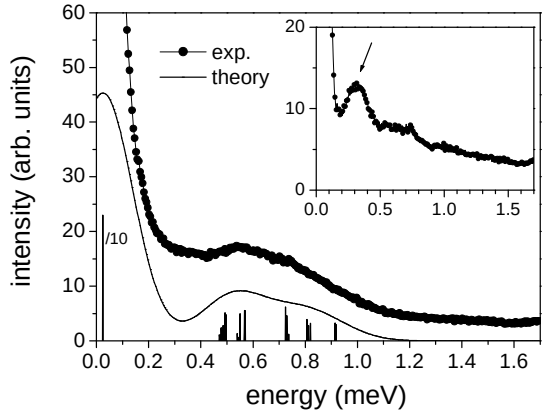


FIG. 2: Inelastic neutron scattering spectrum for $Fe_{30}$. The solid circles show the $T = 65$ mK experimental data of [12] (data were recorded at OSIRIS with final neutron energy $E_f$ =1.845 meV and integrated over the range $Q = 0.9{-}1.8$ Å$^{-1}$). The solid line and the vertical lines represent the $T = 0$ scattering intensity as calculated from SLMFT ($J = -0.125$ meV; the solid line was obtained by a convolution with a Gaussian of linewidth 0.27 meV). The inset displays the experimental data after substraction of the theoretical curve. The arrow marks intensity not accounted for by the theory.

in more detail. The powder INS intensity integrated over $Q = 0.9{-}1.8$ Å$^{-1}$ was calculated from the wavefunctions obtained by SLMFT using the formulas of [24] and the crystallographic Fe positions of $Fe_{30}$.[9] The result for $J = -0.125$ meV is shown as the vertical lines in Fig. 2. Deconvolution of this spectrum with a Gaussian of width 0.27 meV produced the solid curve shown in Fig. 2. The theoretical spectrum exhibits a strong peak at 0.025 meV due to the excitations from the ground state to the $L$-band triplets. It is buried under the large quasielastic contribution and hence not detected in the experiment. Interestingly, the experimental data in the range 0.4 to 1.1 meV is very well reproduced by SLMFT, and the obtained value for $J$ agrees within 7% with the value from magnetic susceptibility, which is a significant improvement over the value obtained with $\hat{H}_{ABC}$. However, in the range 0.2 to 0.4 meV there is additional intensity. It might be described by an "appropriate" background, but considering the resolution function of OSIRIS it should be assigned to a feature not correctly reproduced by the SLMFT. In fact, subtracting the theoretical curve from the experimental data (see inset of Fig. 2) strongly suggests additional scattering from $Fe_{30}$ in this range.

For the discussion, first the conceptional differences between the SWTs and SLMFT are noted. The SWTs start from a symmetry-broken ground state, while SLMFT works with spin levels and is intrinsically spin rotational invariant. Hence, the relation between the states obtained by SWTs on the one side and SLMFT (or exact results) on the other side is not always obvious. For a bipartite system, this is not a big problem. SWTs produce $N$ excitations which relate to $N{-}1$ spin excitations in the $S$=1 sector, i.e., one $L$-band and $N{-}2$ $E$-band triplets (for a system with $S$=0 ground state). There is hence an obvious correspondence. The $L$ band is associated to the (quantized) rotation of the Néel vector, and the $E$ band to the (discrete) spin-wave excitations.[3,4,6,7,25]

For a system with three (or more) sublattices the situation is less clear. The SWTs again produce $N$ excitations, while SLMFT concerns with 3 $L$- and $3(N{-}3)$ $E$-band triplets (for a system with three sublattices and a $S$=0 ground state). Clearly, since there are many more of them, not all of the $E$-band triplets can be spin-wave excitations. This has an important implication. The (zero-temperature) spin-correlation functions are governed by the transition matrix elements $\langle 0|\hat{S}_i^\nu|n\rangle$ ($|0\rangle$ denotes the ground state, $|n\rangle$ the $n$th excited level). Within the sublattice-Hamiltonian approximation therefore only the transitions from the ground state to the triplets of the $L$ and $E$ bands have non-zero intensity, while all other transitions have zero intensity.[6] That is, only the triplets of the $L$ and $E$ bands are relevant. On the other hand, there is no general restriction on the intensities of these transitions. That is, all triplets of the $E$ band are relevant. This is underlined by Fig. 1(b), which reproduces the INS intensity as obtained from SLMFT. Comparison with Fig. 1(a) shows that all parts of the energy spectrum contribute. Since the number of $E$-band triplets considerably exceeds the number of magnons, one necessarily has to conclude that (for three and more sublattices) SWTs are intrinsically not capable of describing the spin dynamics correctly, or completely, respectively.

These points are in favor of SLMFT. However, a comparison of SWTs, SLMFT, and exact results for even-numbered AF rings revealed that SLMFT provides an overall estimate for the spin-wave energies, but fails to reproduce their dispersion (which SWTs of course do).[26] On the other hand, the splitting of the $E$ band in AF finite chains due to the open boundary conditions is well grasped.[27] Apparently, SLMFT accounts for some but not all features of the energy spectrum.

Concerning SLMFT, the studies on the rings and chains showed the following trends. The upper edge of the exact $E$-band spectrum was obtained reasonably well; it was overestimated by about 10%. The dispersion of the magnons, however, was greatly underestimated, yielding very narrow $E$ bands: The exact $E$-band spectrum extends to much lower energies as obtained by SLMFT. These trends could explain the discrepancies in the SLMFT analysis of the $Fe_{30}$ INS data. First, a $J$ value which is 7% too small is consistent with the the trend to overestimate. Second, the scattering intensity in the range 0.2 to 0.4 meV could be associated to magnons, which are at lower energies than calculated because SLMFT does not account for their dispersion.

Concerning the SWTs on the other hand, they were found to reproduce the structure of the $E$ band very well for the rings and chains, but to underestimate energies, i.e., to provide lower bounds for the $E$-band spectrum (the accuracy of LSWT and mLSWT was ca. $-20$%, that of interacting SWT and modified interacting SWT

about −10%). With respect to $Fe_{30}$, Figure 1(a) then would imply that there are no excitations below $3.5|J|$ (besides the $L$-band excitations at very low energies), contradicting an association of the 0.2−0.4 meV intensity to magnons. For the triangular AFM lattice, however, the inclusion of 3rd- and 4th-order boson terms was found recently to result in a substantial suppression of the magnon energies.[28] Since a similar behavior is expected for $Fe_{30}$, the experimental intensity at 0.2−0.4 meV should be indeed associated to magnons, consistent with the above conclusion from SLMFT.

The calculated ground-state energies are $-220.802|J|$, $-220.095|J|$, and $-195|J|$ for the LSWT, mLSWT, and SLMFT, respectively. A recent DMRG calculation yielded $-211(2)|J|$.[10] As observed also for the rings and chains, LSWT yields a lower ground-state energy than mLSWT, while SLMFT is too high. This is linked to the fact that SLMFT does not describe the spin-wave dispersion, and hence misses the related quantum fluctuations. That LSWT and mLSWT, however, yields ground-state energies well below the DMRG result should be considered another hint for the problems of the SWTs.

In conclusion, three theories, LSWT, mLSWT, and SLMFT, have been tested for $Fe_{30}$. The SWTs, as they do not cover all relevant excitations of the $E$ band, are not able to describe the spin dynamics, as measured by INS for instance, correctly. SLMFT was successful to some extend. It reproduces a significant part of the experimental INS spectrum, and yields a $J$ value which is in substantially better agreement with the $J$ value from magnetic susceptibility than that obtained with $\hat{H}_{ABC}$. However, it does not account for all the experimentally observed excitation intensity.

For bipartite systems the combination of the results of SWTs and SLMFT yields a rather good picture of the ground state and low-temperature excitations, even in a quantitative sense.[26] For the three-(and more) sublattice systems, these techniques could be considered useful to some extend for the analysis of experimental data, in particular in the absence of better methods. Conceptionally, however, they all are unsatisfactory. SWTs, on the one side, break the symmetry, which one has then to correct for somehow afterwards. Furthermore, they do not reproduce the complete $E$ band. SLMFT, on the other hand, while overcoming these two issues, is not able to yield the dispersion of the magnons correctly.

In order to further explore the situation for the three-(and more) sublattice systems, a comparison of the SWTs and SLMFT with exact results would be crucial. The spin-1/2 analogue of $Fe_{30}$ can be handled by exact diagonalization.[29] The experience for bipartite systems, however, indicates a breakdown of the $L/E$-band concept for spin-1/2; hence the findings might not be transferable to systems with larger spins.[6] A comparison with the triangular AFM lattice (TL) is thus suggested, as the concepts relevant here and for $Fe_{30}$ are basically identical.[4,20]

Interestingly, the $N$=9 TL is exactly solved by $\hat{H}_{ABC}$, with proper values for $S_A$, $S_B$, and $S_C$ (a similarity in sequence is noted: for bipartite systems $\hat{H}_{AB}$ is exact for the dimer, square, and $N$=8 2D square lattice, for three-sublattice systems $\hat{H}_{ABC}$ is exact for the trimer, octahedron, and $N$=9 TL). Hence, the $L/E$-band concept, and all its implications, is exact here. For rings, which may be regarded as extensions of the dimer and square, the approximation by $\hat{H}_{AB}$ becomes the better the larger $S_i$ and the smaller $N$.[6] For TLs, which may be regarded as extensions of the trimer and $N$=9 TL, one may speculate about a similar trend of the accuracy of $\hat{H}_{ABC}$ with $S_i$.

It would be particularly interesting to study the spin-5/2, $N$=12 TL, because exact diagonalization should be possible (though not on the author's computers). The spin-1/2 case was deeply investigated previously.[4,20] Here it is found that the three $L$-band triplets are slightly split, and that the splitting of the $L$ band increases with increasing $S$. This should be expected to happen also for the spin-5/2 case, and for $Fe_{30}$. This effect could be relevant to explain the observed unusual field dependence of the INS intensity:[12] with increasing field the thermal population of the $L$-band states would change, so that the relative contributions of the $E$-band excitations would change too.

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